

Columnar antiferromagnetic order and spin supersolid phase on the extended Shastry-Sutherland lattice

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We use large scale quantum Monte Carlo simulations to study an extended version of the canonical Shastry-Sutherland model – including additional interaction and exchange anisotropy – over a wide range of Hamiltonian parameters and applied magnetic field. The model is appropriate for describing the low energy properties of some members of the rare earth tetraborides. Working in the limit of large Ising-like exchange anisotropy, we demonstrate the stabilization of columnar antiferromagnetic (CAFM) order in the ground state at zero field and an extended magnetization plateau at $m/m_s = 1/2$ in the presence of an applied longitudinal magnetic field – qualitatively similar to experimentally observed low-temperature phases in ErB_4 . Our results show that for an optimal range of exchange parameters, a spin supersolid ground state is realized over a finite range of applied field between the CAFM phase and the magnetization plateau. The full momentum dependence of the longitudinal and transverse components of the static structure factor is calculated in the spin supersolid phase to demonstrate the simultaneous existence of diagonal and off-diagonal long-range order. Our results will provide crucial guidance in designing further experiments to search for the elusive spin supersolid phase in ErB_4 .

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Ever since Penrose and Onsager [1] speculated on the possible coexistence of diagonal and off-diagonal long range order, supersolid order has been of broad interest within the physics community. Although the initial inspiration for this exotic type of ordering originated from the consideration of the remarkable properties of solid helium, the realization of a supersolid phase of helium remains controversial. [35] On the other hand, theoretical studies of several models of lattice bosons with competing interactions have conclusively established the presence of supersolid phases over extended parameter regimes. [2–6] In this case, the discreteness of the lattice simplifies the process of forming a supersolid, and the bosonic models can potentially be experimentally realized with cold atoms in optical lattices. Concurrently, the realization of BEC of magnons and other novel bosonic phases in quantum magnets provides an alternative route to this elusive state of matter, and the spin analog of the supersolid phase was introduced. [7, 8]. Yet to date, no experimental system has been found that displays unambiguous signs of supersolid ordering. Hence, the identification of realistic systems with supersolid order is a valuable step towards the ultimate realization of this elusive phase.

A key element in the stabilization of a supersolid ground state in many models is geometric frustration, which makes frustrated quantum magnets the natural place to look for the spin supersolid phase. The rare-earth tetraborides (RB_4) are a promising class of quantum magnets for studying the effects of geometric frustration in interacting spin systems. Consisting of weakly coupled layers of magnetic moment carrying R^{3+} ions arranged on the Shastry-Sutherland lattice (SSL), their magnetic properties are well described by appropriate generalizations of the Shastry Sutherland model

(SSM). [9] Several members of this family have been observed to exhibit magnetization plateaus at low temperatures. For example, both ErB_4 and TmB_4 exhibit an extended magnetization plateau at $1/2$ the saturation magnetization. [10, 11] Despite this similarity, however, additional fractional magnetization plateaus have been observed in TmB_4 that are not seen in ErB_4 . Further, the zero-field ground state magnetic structure of these two RB_4 differ significantly. Recent analysis using neutron scattering experiments has determined the low temperature magnetic structure of TmB_4 to be collinear and antiferromagnetic (AFM) with a $\mathbf{Q} = (\pi, \pi)$ ordering of the local moments, [12, 13] i.e. the Ising analog of the Néel state. In contrast, it is well-documented that the magnetic order of the zero-field ground state in ErB_4 , while collinear, is antiferromagnetic only in one direction, i.e. a columnar ordering with either $\mathbf{Q} = (\pi, 0)$ or $\mathbf{Q} = (0, \pi)$. [10, 14]

Previous studies have shown that the low temperature magnetic properties of TmB_4 can be explained by an effective low energy model that is obtained by extending the canonical SSM to incorporate ferromagnetic (FM) exchange with Ising-like anisotropy and additional long range interaction. [15, 16] The additional interaction is instrumental in stabilizing an extended magnetization plateau at $m/m_s = 1/2$, while at the same time eliminating the magnetization plateau at $m/m_s = 1/3$ that is ubiquitous to the canonical SSM in the Ising limit. [17, 18] Similar generalizations are expected to capture the low-energy properties of other members of the RB_4 family. Thus, it is useful to extend the results of this effective model to include magnetization processes that begin from zero-field ground states other than the Néel-like antiferromagnetic state considered so far.

In this Letter, we determine the parameter regime of the effective model for RB_4 for which a columnar antiferromagnetic ordering is stabilized in the absence of an external field. We present large scale quantum Monte Carlo (QMC) simulations of the magnetization process in this regime and demonstrate the appearance of a field-induced spin supersolid phase at magnetizations below half saturation. Arguing that similar factors may drive the stabilization of columnar order in ErB_4 , we identify ErB_4 as a promising candidate for the observation of supersolid order.

Many RB_4 are well-suited to numeric investigation of their low-temperature magnetic properties. The high total angular momentum J of the R^{3+} ions, combined with a large easy-axis single-ion anisotropy, allows us to construct a simple effective model of the low energy states. Briefly, the large easy-axis single-ion anisotropy, D , splits the local Hilbert space at each site into degenerate doublets – the successive doublets are separated by energy gaps $\sim D$. To a first approximation, then, the low-energy magnetic interactions are captured by an Ising model comprised of the maximal- J doublet. Higher order processes lead to weak FM (AFM) transverse exchange interactions between neighboring sites for integer (half-odd integer) values of J , and thus the XXZ model with Ising-like exchange anisotropy becomes the preferred model to study the low-energy magnetic properties of ErB_4 and TmB_4 , both of which possess a large easy-axis single-ion anisotropy. [19] In this work, we restrict ourselves to the case of FM transverse exchange interactions, where the model is free of the negative sign problem and thereby directly amenable to QMC simulation.

The above model can be described by the Hamiltonian

$$\mathcal{H} = \sum_{\alpha=1}^3 \sum_{\langle ij \rangle_{\alpha}} [-|J_{\alpha}\Delta| (S_i^x S_j^x + S_i^y S_j^y) + J_{\alpha} S_i^z S_j^z] - h_z \sum_i S_i^z, \quad (1)$$

where the summation is over all bond types α of strength J_{α} and their associated bonds $\langle ij \rangle_{\alpha}$ acting on sites i and j of the extended SSL depicted in Fig. 1(a). Here, in addition to the canonical interactions J_1 and J_2 of the SSM, we have included an additional interaction J_3 along the diagonals of the “empty” plaquettes (i.e. those missing the J_2 interaction). We work in the experimentally relevant limit of strong Ising-like exchange anisotropy, $\Delta \ll 1$, and consider the effect of an external magnetic field h_z . Since the crystal structure of these materials is such that the bonds of the canonical SSL are expected to be of roughly equal strength, in what follows we set $J_1 = J_2 = 1$, and all parameters are given in these units.

To characterize the different phases of \mathcal{H} , longitudinal and transverse components of the static structure factor

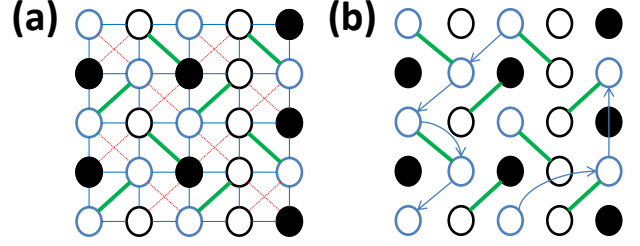


FIG. 1: (Color online) Ising ground state structure of the half plateau state for antiferromagnetic J_3 . Black and white circles represent down and up spins, respectively, while blue outlines indicate spins that can be flipped with minimal energy cost. The bonds J_1 , J_2 , and J_3 (see text) are illustrated in (a) as solid blue, thick green, and dotted red lines, respectively. In (b) the hopping processes in the spin supersolid phase are illustrated (J_3 bonds are omitted for clarity).

are defined as

$$S^{+-}(\mathbf{k}) = \frac{1}{N} \sum_{i,j} \langle S_i^+ S_j^- + S_i^- S_j^+ \rangle e^{i\mathbf{k} \cdot (\mathbf{r}_i - \mathbf{r}_j)},$$

$$S^{zz}(\mathbf{k}) = \frac{1}{N} \sum_{i,j} \langle S_i^z S_j^z \rangle e^{i\mathbf{k} \cdot (\mathbf{r}_i - \mathbf{r}_j)}. \quad (2)$$

In the S^z basis, these serve as measures of off-diagonal and diagonal order, respectively. As the uniform magnetization per site is simply $m_z = \sum_j \langle S_j^z \rangle / N$, here we note that $S^{zz}(\mathbf{0}) = N \langle m_z^2 \rangle$. Further, we define staggered and columnar magnetizations as $\langle m_s^2 \rangle = S^{zz}(\pi, \pi) / N$ and $\langle m_c^2 \rangle = [S^{zz}(0, \pi) + S^{zz}(\pi, 0)] / N$, respectively.

QMC simulations are performed using the stochastic series expansion algorithm with directed loop updates. [20] The correlations $\langle S_i^+ S_j^- + S_i^- S_j^+ \rangle$ and $\langle S_i^z S_j^z \rangle$ can be straightforwardly computed within this method [21, 22]. A useful observable in characterizing the ground state phases in numerical simulations is the spin stiffness, ρ_s , defined as the response to a twist in the boundary conditions. [23] In simulations that sample multiple winding number sectors, the evaluation of the stiffness simplifies to calculating the winding number of the world lines, leading to the expression $\rho_s = (w_x^2 + w_y^2) / 4\beta$ in two dimensions, [24] where w_x and w_y are the total winding in the x and y directions.

Let us begin by considering the ground-state phase diagram of \mathcal{H} in the Ising limit, $\Delta = 0$. In this limit, we can construct the various possible ground states by hand. By comparing their respective energies, the phase diagram in the $h_z - J_3$ plane is determined (see solid black lines and labeled phases in Fig. 2). This will provide a background of classically ordered magnetic states upon which we shall study the effect of quantum fluctuations. At zero field the ground state is an Ising antiferromagnet with either staggered ($\mathbf{Q} = (\pi, \pi)$) or columnar ($\mathbf{Q} = (\pi, 0)$ or $\mathbf{Q} = (0, \pi)$) order. We will refer to these states as SAFM

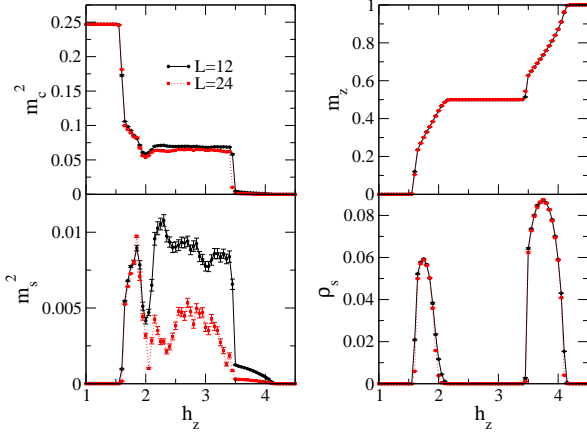


FIG. 3: (Color online) Behavior of the columnar (m_c^2), staggered (m_s^2), and uniform (m_z) magnetizations, along with the spin stiffness (ρ_s), for the complete magnetization process at $\Delta = 0.10$, $J_3 = 1.25$, and inverse temperature $\beta = 64$. The sequence of phases with increasing magnetic field is columnar, supersolid, half plateau, superfluid, and fully polarized.

in S^{zz} develops spontaneously at $\mathbf{k} = (\pi, \pi)$ in the SS phase through a quantum order-by-disorder process that we discuss below.

The mechanism for superflow in the SS phase can further explain the observed features of the transverse structure factor. Since the SS phase occurs at magnetizations just below half saturation, the mechanism for supersolid formation can be determined by considering the hopping processes available when the HP state is doped with down spins. The lowest potential energy doping sites in the HP phase for $J_3 > 1$ are depicted by white circles with blue outlines in Fig. 1. It is clear to see that down spins at these sites can easily delocalize by first order processes when the phase of AFM stripes is conserved. In contrast, along the domain wall type structure of out-of-phase stripes, only second order hopping processes are allowed. These two hopping processes are depicted in Fig. 1(b). Interestingly, since the domain walls impart no potential energy gain, the total energy will be lower with the first order hopping processes of phase-matched stripes. Thus, the degeneracy of the HP state is partially lifted by quantum fluctuations in the SS phase, leading to a higher degree of diagonal order. This effect is expected to grow stronger with increasing J_3 values, and is an example of a quantum “order by disorder” process. [27–30]

The model Hamiltonian \mathcal{H} can be mapped onto a system of hardcore bosons with nearest-neighbor hopping $t = -|J_\alpha \Delta|/2$ and repulsion $V = J_\alpha$. For such a system on the triangular lattice it has been shown [3–6] that a supersolid phase is stabilized for weak hopping $t \ll V$ or $\Delta \ll 1$. Similarly, a square lattice model of hardcore bosons with next-nearest-neighbor repulsion (similar to our J_2 and J_3 terms) is known to possess both colum-

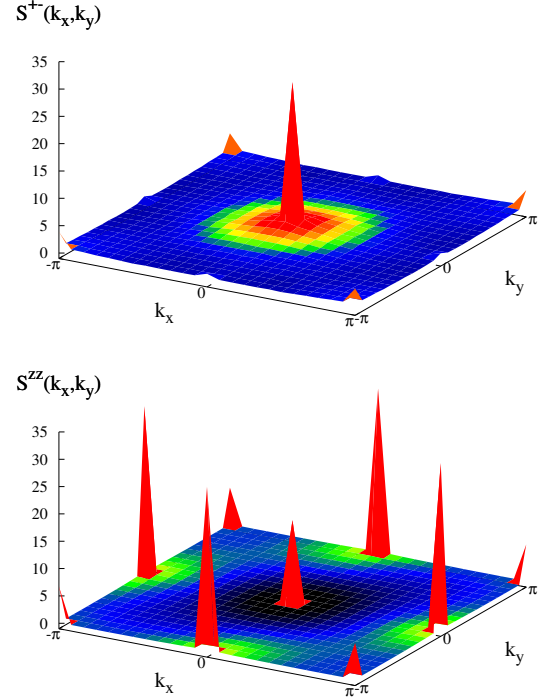


FIG. 4: (Color online) Transverse and longitudinal structure factors in the spin supersolid phase, with $S^{+-}(\mathbf{k})$ and $S^{zz}(\mathbf{k})$ as defined in Eq. 2. Data are taken from a 24x24 lattice at inverse temperature $\beta = 100$. The free parameters are set to $\Delta = 0.10$, $h_z = 1.75$ and $J_3 = 1.25$ to assure that we are well within the spin supersolid phase as outlined in Fig. 2.

nar order and supersolid phases (see [31] and references therein). While no such supersolid state has been found for hardcore bosons on the canonical SSL, [32] we have shown that the addition of antiferromagnetic J_3 bonds allows for the formation of a supersolid phase on the extended SSL.

Let us now turn to the experimentally determined magnetization process of the rare-earth tetraboride ErB_4 . Our calculations have shown that it is possible to obtain the observed magnetization process *starting from a zero-field columnar antiferromagnetic state*. The theoretically calculated magnetization process compares well not only qualitatively, but also quantitatively, as reflected by the extent of the HP state ($\Delta h_{1/2}/h_s \sim (3.5-2)/4$ or 38% in our model vs. $\Delta h_{1/2}/h_s \sim (4-2)/5$ or 40% in Ref. [10]). Additionally, this magnetization process may contain a spin supersolid phase for magnetizations below half saturation, as determined by our simulations of an effective model with FM transverse exchange interactions. While the total angular momentum $J = 15/2$ for ErB_4 actually leads to AFM transverse exchange interactions, we believe that a similar mechanism could lead to a spin supersolid phase with AFM transverse order. Interestingly, such an ordering in the effective transverse spin degrees

of freedom would correspond to *antiferromagnetic* order of the total angular momentum. [33]

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